**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "The hygroscopicity parameter (κ) of ambient organic aerosol at a field site subject to biogenic and anthropogenic influences: Relationship to degree of aerosol oxidation" *by* R. Y.-W. Chang et al.

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The topic of this study is highly relevant, field measurements were conducted with state-of-the-art experimental techniques and data analysis as well as manuscript preparation have been done with care. The pretended key result of this study is a correlation between O/C and CCN activity of the organic aerosol fraction (expressed as "kappa_org"). However, no conclusive evidence for this relationship is given, instead the result is merely imposed by the assumptions of the chosen parametrization, as will be detailed below. The data presented in this

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manuscript just show that kappa_org is larger than zero, i.e. the organic fraction enhances CCN activity beyond its mere contribution to particle size.

It is possible that the data set of this study also allows establishing a link between O/C and kappa_org. However, showing this definitely needs additional data analysis. A major revision of this manuscript is required due to the vital gap in the line of argument.

p. 25335 and Figure 1: The results with the approach presented on p. 25335 and in Fig. 1 just show that the campaign mean kappa_org is larger than zero, while no conclusive evidence of a correlation between the ratio of OOA to HOA and kappa_org is given. The kappa of the oxygenated organic fraction (kappa_ox) will always end up positive if the unoxygenated fraction is assumed to be non-hygroscopic (kappa_unox=0) and the campaign mean kappa is larger than zero at the same time, independent of the true cause for kappa_org>0. Similarly one would end up with kappa_unox>0 if kappa_ox was assumed to be zero, while varying kappa_unox until best closure was achieved. The analysis presented in this paper just shows that the assumption kappa_org=eps_ox-*kappa_ox gives better CCN predictions than the assumption kappa_org=0. The latter assumption has previously been shown to be invalid in several environments (see e.g. references in this paper). The key thing to be shown here is that the assumption kappa_org=eps_ox * kappa_ox leads to significantly better individual CCN predictions compared to alternative approaches such as kappa_org=constant (0.14 according to p. 25342, line 16) or the counter-intuitive assumption kappa_org=eps_unox * kappa_unox. Without doing so, no evidence of a causal link between the ratio of OOA to HOA and kappa_org is given. A further approach to be tested is leaving both kappa_unox and kappa_ox as free parameters in the optimization, similar to the approach in the study by Raatikainen et al. (2009), which investigated the link between different AMS-PMF-derived OA subfractions and hygroscopicity at subsaturated RH, volatility,

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etc.

We thank the reviewer for his helpful comments. We have extensively re-written the paper to make the analysis techniques, and in particular the associated assumptions, clearer.

In particular, we fully agree that we have not demonstrated (in the original manuscript) that κ_{org} correlates with the degree of oxygenation of the aerosol, whether with the O/C or with the oxygenated factor. Although we thought the assumptions that we made in the analysis were clear in the original manuscript, we have now made these statements explicitly in the Abstract and a number of other parts of the paper to avoid confusion.

To briefly summarize, we make an assumption that the hygroscopicity of the organic component varies with its degree of oxygenation. Specifically, we assume that the oxygenated factors contribute to the hygroscopicity of the aerosol, and that the unoxygenated factor does not, whether due to its solubility, as suggested by Petters et al. (2009), or its intrinsic hygroscopicity. Under this assumption, we then determine how our field data inform us about the hygroscopicity of the oxygenated fraction.

Although attempting to fit for the κ of both the oxygenated and unoxygenated components would have been the ideal approach, the uncertainties in our measurements were too great to evaluate the difference between such sets of κs. As such, we constrained our model by assuming that the unoxygenated (HOA) component of the organic aerosol contributed minimally to the aerosol hygroscopicity. This is a reasonable assumption because HOA is thought to be hydrocarbon-like (Zhang et al., 2005) and aliphatic in nature, and in general, these types of compounds have not been found to be CCN-active in laboratory experiments (e.g. Raymond and Pandis, 2002; Pradeep Kumar et al., 2003). In addition, ambient measurements of particles at locations in which HOA is high, especially at the smaller sizes, have shown that the organic component does not appear to contribute to the CCN-activity (Cubison et al., 2008; Quinn et al.,

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2008). Based on these published results, we felt that the assumption that the unoxygenated component was non-hygroscopic was reasonable.

In contrast to the HOA factor, the OOA factor, which makes up most of the oxygenated component of the organic aerosol, is thought to be characteristic of highly processed organic aerosols (Zhang et al., 2005) formed from secondary processes (Zhang et al., 2007). Again, laboratory experiments of compounds that are formed from secondary processes such as glutamic, glutaric, pinonic and norpinic acids have found that these aerosols are moderately CCN-active (Raymond and Pandis, 2002) and smog chamber studies of gaseous organic precursors with ozone or OH have also found that the organic aerosols formed are moderately CCN-active (e.g. Duplissy et al., 2008). These findings support the idea that the oxygenated component of the organic aerosol is CCN-active and that it is not unreasonable to assume that it contributes to the overall aerosol hygroscopicity. As such, we believe that assuming that the unoxygenated component is non-hygroscopic and that the oxygenated component is hygroscopic is consistent with what is known from the literature. Under this assumption, the goal of the paper is to extract only one piece of information, i.e. the hygroscopicity of the oxygenated component of the organic aerosol. This discussion is elaborated upon greatly in the revised manuscript (Sect. 3.3).

p. 25339, first paragraph of Section 4.2: This approach just shows that kappa_org>0, while it does not give evidence of a correlation between O/C and kappa_org, similar to the previous comment. Starting of with the presumption that kappa_org correlates with O/C (i.e. assuming kappa_org = a * (O/C)) will always result in a positive value for a, if the campaign mean kappa_org is larger than zero. With the "opposite" assumption that kappa_org = b*(C/O), one would end up with something like b ~ 1/a I guess, which is also a positive value. The key thing to be shown with the data set of this study is not that the assumption kappa_org = a * (O/C) performs better than the assumption kappa_org=0. Instead

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it has to be shown that kappa_org = a * (O/C) gives significantly better CCN predictions for the individual data points compared to alternative assumptions such as kappa_org=constant (optimized of course) or kappa_org = b * (C/O).

The point the Reviewer is making here is similar to his first, i.e. in the original manuscript he is stating that we do not show that a correlation exists between κ_{org} and O/C. We agree. As stated in the original version of the paper, the functional form of Eq. 5 and 6 is assumed.

To address this point, we now include a graph (new Fig. 4) of inferred κ_{org} vs O/C in Sect. 4.1.2 in the revised manuscript, where we have averaged κ_{org} values for different O/C ratios. The scatter in this plot was the reason that we originally did not include it in the manuscript, i.e. we are hesitant to derive a functional relationship from these data preferring, instead, to propose Eq. 5 based on chemical intuition (i.e. that the κ_{org} value would converge to zero for O/C of zero and to higher values for higher O/C). Figure 4 illustrates that the uncertainties are quite large for the average inferred κ_{org}. Nevertheless, disregarding the uncertainties for the time being, there is a positive correlation between κ_{org} and O/C over our relatively narrow range of measurements. As noted in the response to Reviewer 3, the data points in Fig. 4 are quite similar to those of Jimenez et al. (2009) for the κ_{org} values derived from both lab and field data for subsaturated conditions. We also include in the paper the functional form of the correlation that is presented in Fig. 4. However, given the uncertainties involved, we do not consider our data to be a strong test of the inherent assumptions in our analysis, i.e. that the oxygenated component of the organic aerosol drives the hygroscopicity in a linear manner. Furthermore, in Sect. 4.4 in the revised text, we point out that if a constant κ_{org} of 0.15 is used in our analysis, the organic-rich particles tend to be slightly overpredicted compared to the inorganic-rich particles.

Section 4.1.2, Table 1 and Fig. 5: Deriving kappa_ox and kappa_unox separately

for different time periods is a step towards testing the generality of the "postulated" relationship kappa_org = eps_ox*0.20. However, my interpretation of the results is opposite from the author's interpretation. The fact that the retrieved kappa_ox is massively different for one out of just three periods rather indicates that kappa_org is mainly controlled by other factors than the just ratio of HOA to OOA, and that the above relationship cannot be recommended for general use, as it is put in abstract and conclusions.

Although only representing 11% of the full study, these time periods to which the reviewer is referring do exhibit different CCN behaviour than for the rest of the periods examined. So the reviewer has a fair comment. Although present in the original manuscript, we now stress in the Conclusions that not all time periods adhere to the average results for the entire study.

p. 25339, line 24 - p. 25340, line 6: This whole paragraph including Table 2 is pure speculation and has to be removed. The kappa values of the different PMF factors were not independently derived, they are just calculated under the assumption that the relationship inferred between k_org and O/C is generally applicable. Above I argued that validity of this relationship has not been proven and the authors themselves state that "The oxygenated factors were grouped together because our model was not sensitive enough to elucidate separate hygroscopicities for each factor." on p. 25333 of their manuscript.

We have removed this paragraph and the associated parts of the Table from the paper.

Minor comments

p.25327, lines 2-5: In the mean time Jimenez et al. (2009) have published a paper

in which the O/C of the organic aerosol component of laboratory generated and ambient aerosols is linked to the hygroscopicity at subsaturated relative humidity (RH). Another study by Raatikainen et al. (2009) investigates the link between different AMS- PMF-derived OA subfractions and hygroscopicity at subsaturated RH, volatility, etc.

References to these papers have been made in the revised manuscript in the Introduction.

p. 25329, lines 3-7: Please cite the source of thermodynamic data used for calculating the CCN properties of calibration substance ammonium sulfate. Did you assume a kappa value of 0.61 as mentioned later in the manuscript for the closure calculations? The kappa value of pure ammonium sulfate at the point of CCN activation depends on the supersaturation (SS). It changes from 0.65 at SS=0.1% to 0.55 at SS=1.0% and it is 0.59 at SS=0.42.

Thank you for bringing this to our attention. A value of 0.59, as calculated from activity coefficients from Clegg et al. (1996) at 0.42% supersaturation, is now used and carried through in the analysis. This is now explained in the Sect. 3.1 of the revised manuscript. [Note: The original calculation used 0.61 both to calibrate the effective supersaturation of the chamber and for the data analysis, so the results do not significantly change by using the (correct) value of 0.59.]

p. 25330, lines 12-20: I recommend using the terminology of Jimenez et al. (2009) for the PMF factors. "low volatility OOA" and "semi-volatile OOA" are more meaningful terms than OOA-1 and OOA-2.

We agree that the LV-OOA and SV-OOA terminology is more meaningful in terms of understanding the properties of the organic component. However, tracer correlations suggest that the time series in this study are primarily driven by factors other than volatility, despite the fact that the OOA-1 and OOA-2 factor mass spectra are similar to those of LV-OOA and SV-OOA, respectively (Slowik et al., 2010). As such, we continue to use the more general OOA-1/OOA-2 terminology in the manuscript, but we include the newer terms after each entry, e.g. OOA-1 (LV-OOA). This also allows this manuscript to be consistent with the previously-published papers from this study. This is now mentioned in the paper (Sect. 2.4).

p. 25332, lines 14-19: Is the simplification to approximate ammonium nitrate by ammonium sulphate needed. The calculations would hardly become more complex with distinguishing them. Furthermore, if this simplification is maintained, then the associated uncertainty in the derived kappa of the organic aerosol fraction has to be added to the (nice) sensitivity analysis presented in Section 4.3). Furthermore, please make sure that the kappa value of ammonium nitrate at the point of activation at SS=0.42% is indeed 0.67 as stated.

The κ for ammonium nitrate is in fact 0.72 at 0.42% supersaturation (thermodynamic data from Clegg et al. (1998); Wexler and Clegg (2002)). This was an oversight on our part. It is true that calculating ammonium nitrate separately is not complicated, however, we felt that it was easier to simplify the inorganic component. The uncertainty associated with this assumption is not great and has been added to the Uncertainties section.

p. 25333, line 11: Duplissy et al. (2008) have shown that O/C has a strong influence on hygroscopicity of alpha-pinene SOA at RH=95%. The effect of varying O/C, caused by varying precursor concentration, on CCN activation

of alpha-pinene SOA was not shown in Duplissy's study, instead it was just inferred from the behavior at RH=95% and the presumption that CCN activity will change similarly (CCN data very only available for high O/C from low precursor concentration experiments). However, a follow up study by Juranyi et al. (2009) showed that varying O/C of alpha-pinene SOA has virtually no influence on CCN activation, while the strong effect on hygroscopicity at lower RH was confirmed. Petters et al. (2009b) provides theoretical arguments that for this difference between "low" RH and very dilute solutions at the point of CCN activation. See also references therein for further experimental studies.

This statement refers to the reported increase of κ from 0.09 to 0.12 in paragraph 6 and the open symbols in the middle panel of Fig. 2 of the Duplissy et al. (2008) paper. While it is true that Petters et al. (2009) discuss the difference between kappa at super- and subsaturated conditions, their companion paper, Wex et al. (2009) did measure a lower κ under supersaturated conditions for their SOA generated in the presence of butanol (an OH scavenger) compared to experiments without butanol, which were inferred in their paper to be more oxidized (refer to Fig. 8 of their paper).

This paragraph has been moved to the new section in the revised manuscript that discusses the relationship between degree of oxygenation and hygroscopicity (Sect. 3.3) and now reads:

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 "This is supported by laboratory studies that have found
that oleic acid aerosols that have undergone ozonolysis have
become somewhat more CCN-activity (Broekhuizen et al., 2004;
Shilling et al., 2007) and chamber studies that show that
ageing organic aerosols can also increase their CCN-activity to
varying amounts (Duplissy et al., 2008; Petters et al., 2006;
Wex et al., 2009).  We do note that there is also some evidence
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that the hygroscopicity of secondary organic aerosols under
supersaturated conditions in chamber studies do not vary with
ageing (Prenni et al., 2007; Jurányi et al., 2009)."
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 p. 25333, lines 14-15: According to the arguments brought up in Petters et al. (2009b) one does not necessarily expect a strong dependence of CCN activity (CCN-derived kappa_org) on O/C from a theoretical point of view. Molecular weight gains on importance compared to O/C when it comes to very dilute solutions, in contrast to the influence of O/C on hygroscopicity at lower water activity.

This sentence has been changed to:

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 "Under these assumptions, there is the expectation that the
aerosol's hygroscopicity will increase with the degree of
oxygenation, although this relationship would intrinsically
arise from factors such as molecular weight or solubility, that
would affect hygroscopicity and be related to the oxygenation
of the organic components."
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and it is further discussed in the new Sect. 3.3 and Sect. 4.1.2.

 p. 25333, line 26 - p. 25334, line 2: Adipic acid - which has a relatively low solubility - may not be most representative for OOA. Cross et al. (2007) have reported experimental density data.

The organic component has been made more consistent throughout the paper and is

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now treated as two components, oxygenated and unoxygenated, for all the calculations including the density. The oxygenated component, in particular the OOA component, is thought to be similar to secondary organic aerosol (SOA). As such, a density of 1500 kg m^{-3} (measured for monoterpene SOA by Kostenidou et al. (2007)) was used for the oxygenated component. On the other hand, the unoxygenated component, i.e. HOA, is thought to be composed of, among other components, lubricating oil, which has a density of 900 kg m^{-3} (e.g. Cylinder Lube 1000 from NOCO Energy Corp.). As such, this was the density used for the unoxygenated component. In regards to the solubility of the oxygenated component, increasing the solubility by ten times does not affect our final results, suggesting that at activation, it has completely dissolved.

p. 25336, lines 4-7: An average ratio (R_CCN) of 1.20 between predicted and measured CCN concentration seems inconsistent with a fitted slope of 1.02 between the same two quantities. First, is the distribution of individual R_CCN values rather normal or lognormal? In the latter case it is better to report the geometric mean instead of the arithmetic mean of the R_CCN values. Second, Fig. 2 shows that the R_CCN are biases towards values >1 mainly at medium and low CCN concentrations, while they are near unity at the highest CCN concentrations. Fitting a line with axis intercept at the origin is most sensitive to the highest CCN concentrations unless a weighting is introduced.

The distribution was indeed lognormal, and the geometric mean was lower than the arithmetic mean (1.18 and 1.25 for the organic- and inorganic-rich particles, respectively). However, it is still not enough to account for the difference. In an attempt to reduce the weighting of the higher data points, the logarithm of the data points were taken and then linearly fit, but the slope only increased slightly to 1.03. If the data points are weighted by their estimated error (20% based on the uncertainties of the measured CCN concentrations) and the intercept set to zero, then the slope can

be increased to 1.10 (1.05 if the intercept is not forced through zero), which halves the discrepancy with the value obtained from R_{CCN}, but still does not explain all of it. We agree that these values are inconsistent but we cannot reconcile them. Any suggestions that you may have would be welcome. This discussion has been included in Sect. 4.1.1 of the revised manuscript.

p. 25338, lines 3-16: This is largely a repetition of text already given on page 25335. Please try to reduce redundancy.

Section 3 has been restructured to reduce redundancy and to make the text more clear.

p. 25341, line 16-20: The studies by Juranyi et al. (2009) and Petters et al. (2009b) indicate that O/C has little influence on CCN activity of organic compounds.

p. 25341, line 27ff: Ensemble mean molecular weight is also a key factor for CCN activity of organics, not just O/C.

These issues are discussed in the newly added Sect. 3.3 and sections of this paragraph have been modified as such:

"Results from laboratory studies are inconclusive about the relationship between organic oxygenation and hygroscopicity under supersaturated conditions. Unoxygenated organic aerosols that are non-hygroscopic such as oleic acid, stearic acid and bis-2-ethylhexyl sebacate, can be heterogeneously oxidised such that they become moderately hygroscopic (Petters et al., 2006;

Broekhuizen et al., 2004; Shilling et al., 2007; George et al.,
2009). A smog chamber study by Wex et al. (2009) also showed
that the hygroscopicity of SOA increased in the absence of an
OH scavenger, which the authors suggested was due to increased
oxidation in the organic aerosol. Finally, Duplissy et al.
(2008) also showed that the hygroscopicity increased slightly
throughout the photoxidation of α-pinene. However, there
have also been several studies of SOA in smog chambers that
have not observed any dependence of hygroscopicity on degree
of oxygenation, whether by varying precursor concentrations
that should result in varying oxygenation in the final aerosol
(Jurányi et al., 2009), or by monitoring the hygroscopicity
throughout a reaction (Prenni et al., 2007). As such, the
dependence of hygroscopicity on the degree of oxygenation from
laboratory studies is unclear. However, the applicability of
these lab experiments to ambient conditions has not been fully
confirmed, given that chamber-generated SOA tend to be less
oxygenated than the OOA-1 (LV-OOA) component found in highly
processed ambient aerosols (Ng et al., 2009). Although the
focus of this work is on supersaturated conditions, and it is
unclear whether such κ parameters are fully the same as those
derived from subsaturated conditions, we do note that κ for
organic aerosols measured under subsaturated conditions have
recently been reported to scale with the O/C of the aerosol
(Jimenez et al., 2009)."

p. 25343, lines 1-4: As detailed above I do not at all agree with this summary statement.

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This statement has been deleted.

p. 25343, conclusions, lines 11-12: "most of the specific time periods for the study". Call 2 out of 3 "most" is audacious (see also corresponding major comment).

The wording has been changed in the Conclusions.

Technical corrections:

p. 25332, line 2: What do you mean with "initially"? Isn't it simpler to directly state that you assume surface tension of pure water in all closure calculations.

We agree. The "initially" was extraneous and has been removed.

References

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